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Title: Nuclear Forensics and Radiochemistry: Reaction Networks

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Nuclear Forensics and Radiochemistry: Reaction Networks

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Abstract:
In the intense neutron flux of a nuclear explosion the production of isotopes may occur through successive neutron induced reactions. The pathway to these isotopes illustrates both the complexity of the problem and the need for high quality nuclear data. The growth and decay of radioactive isotopes can follow a similarly complex network. The Bateman equation will be described and modified to apply to the transmutation of isotopes in a high flux reactor. A alternative model of growth and decay, the GD code, that can be applied to fission products will also be described.

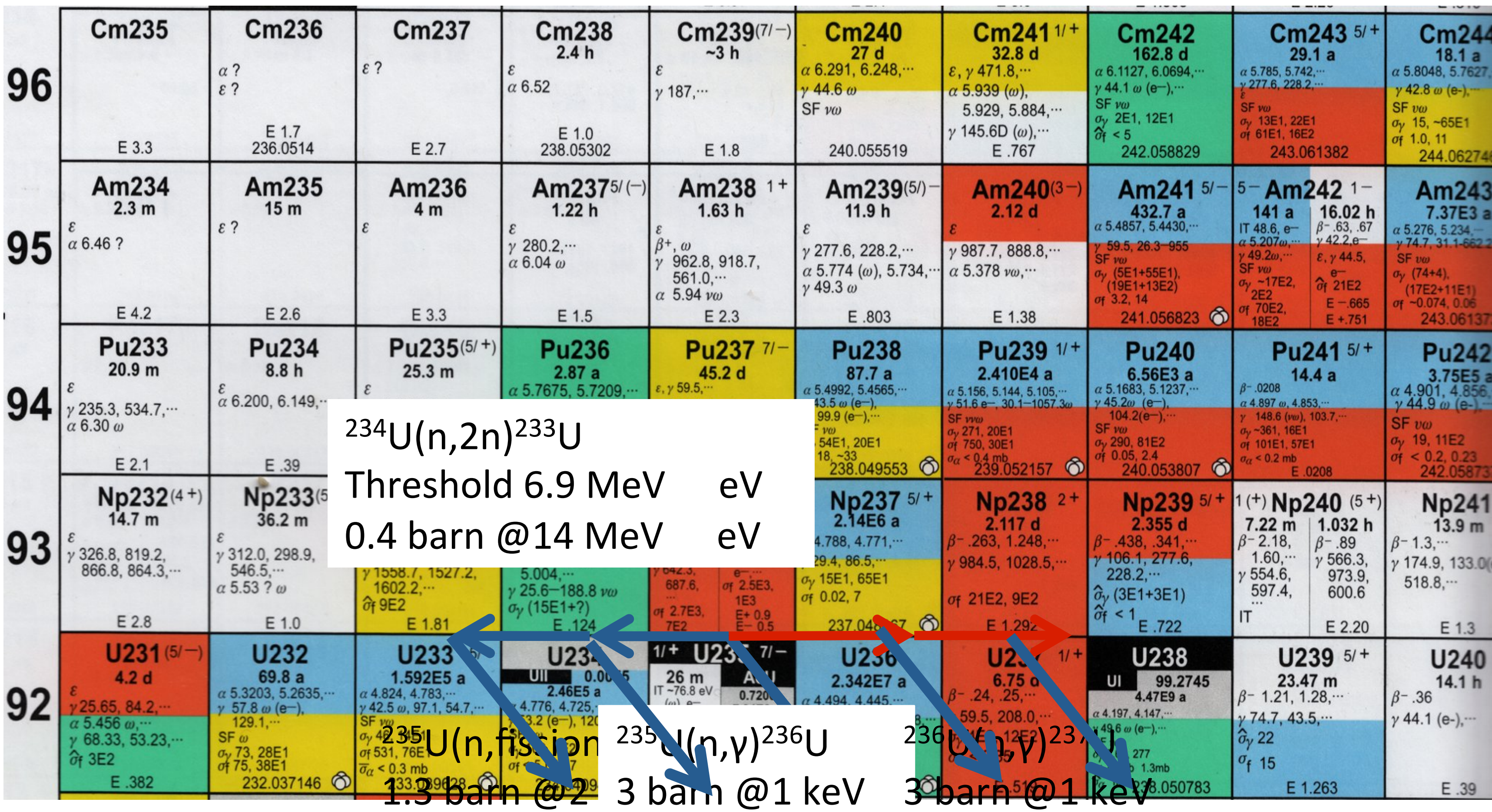
Nuclear Forensics and Radiochemistry: Reaction Networks

Lecture 5

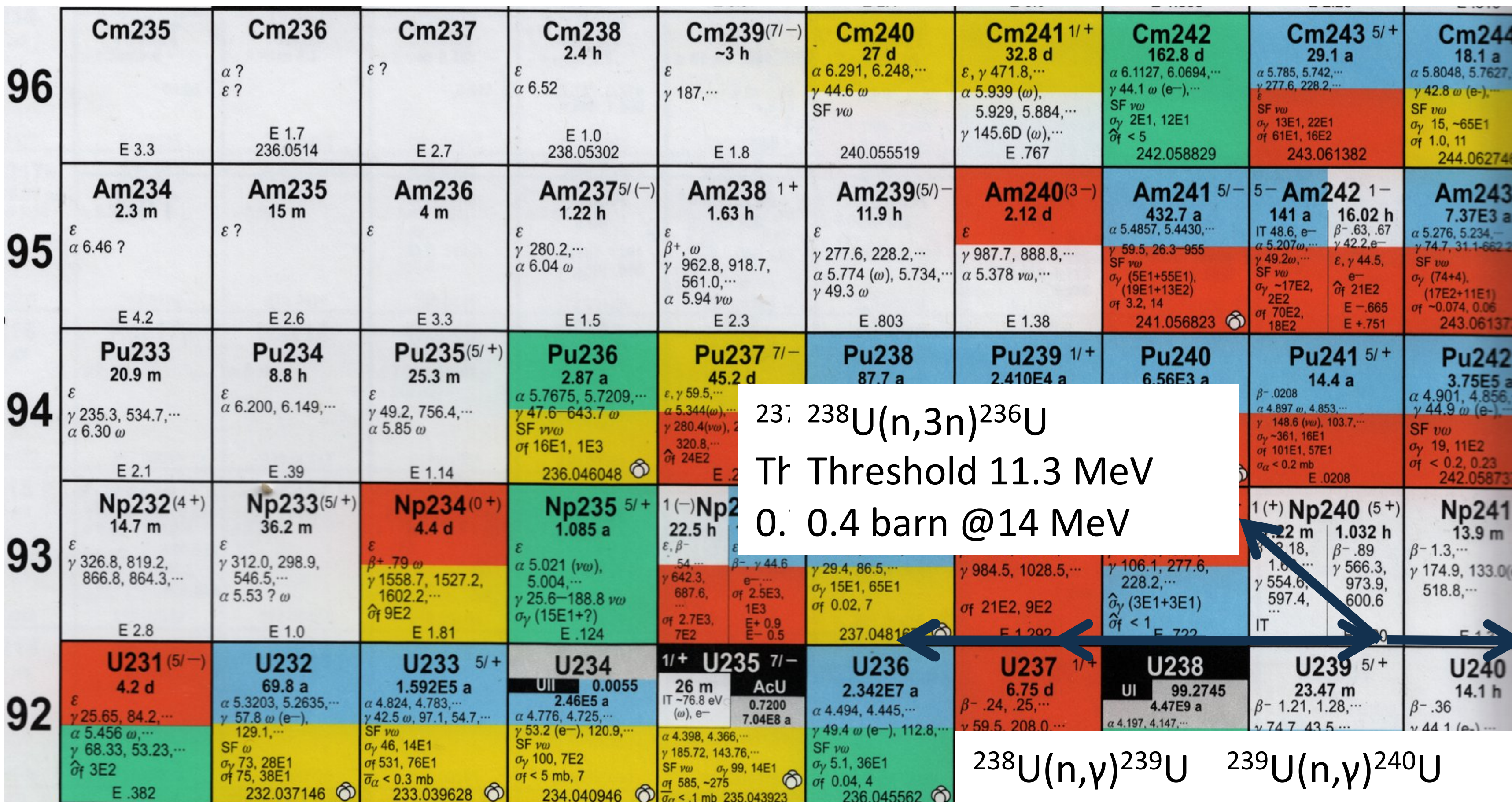
Reaction Networks

- A nuclear explosion produces an enormous flux of neutrons.
  - A neutron yield on the order of a mole (6.02E23).
  - The chain reaction is finished in a short time, of the order of tens of nano-seconds.
  - The volume of burning fuel is relatively small.
- Multiple successive neutron induced reactions can occur.
- Activation of materials by successive reactions are prominent near the fuel.

Neutron Reactions with Actinides (Uranium)



Neutron Reactions with Actinides (Uranium)

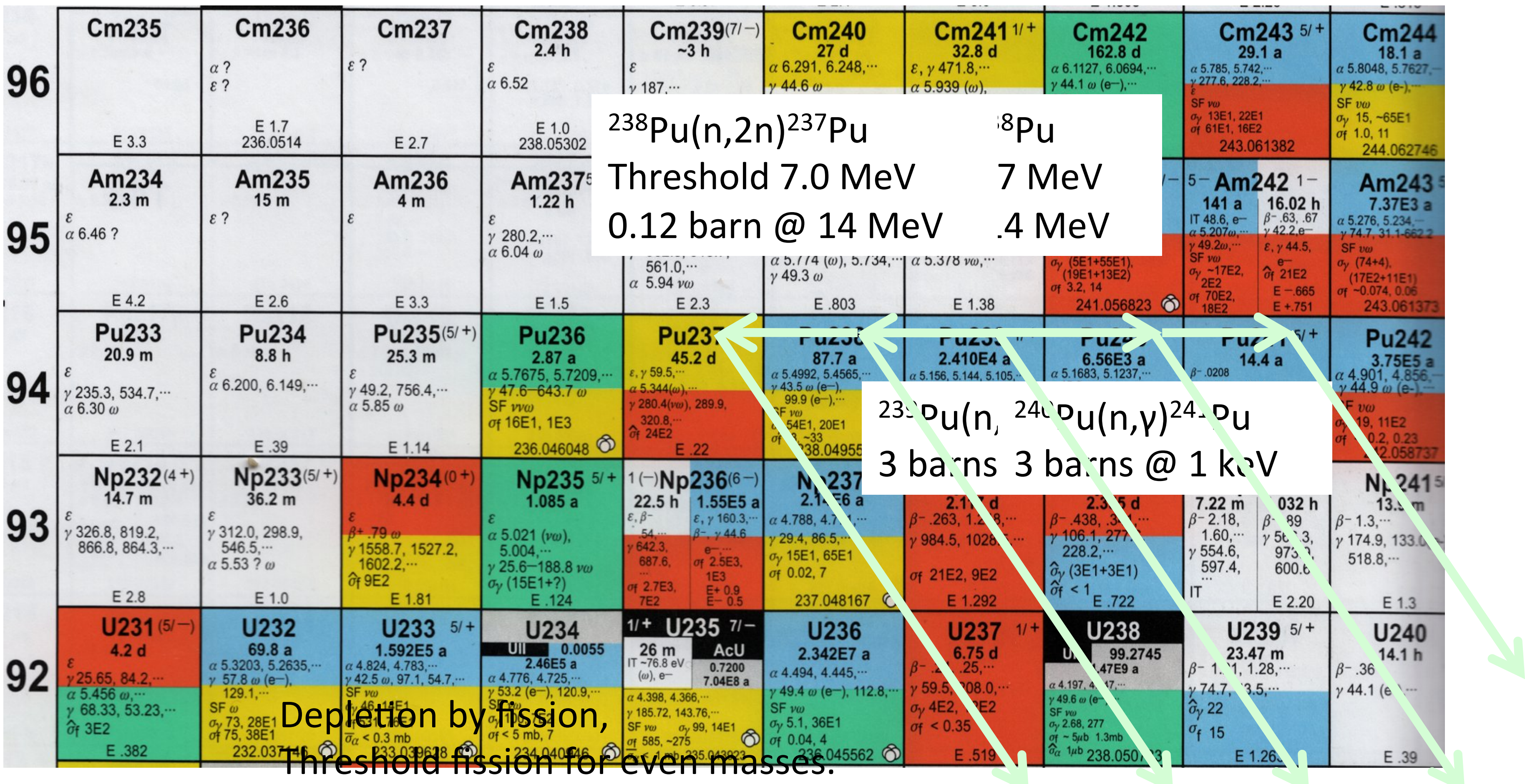


$^{238}U(n,3n)^{236}U$
Threshold 11.3 MeV
0.4 barn @14 MeV

$^{238}U(n,\gamma)^{239}U$
0.4 barn @1 k

$^{239}U(n,\gamma)^{240}U$
3.0 barn @1 keV

Neutron Reactions with Actinides (Plutonium)



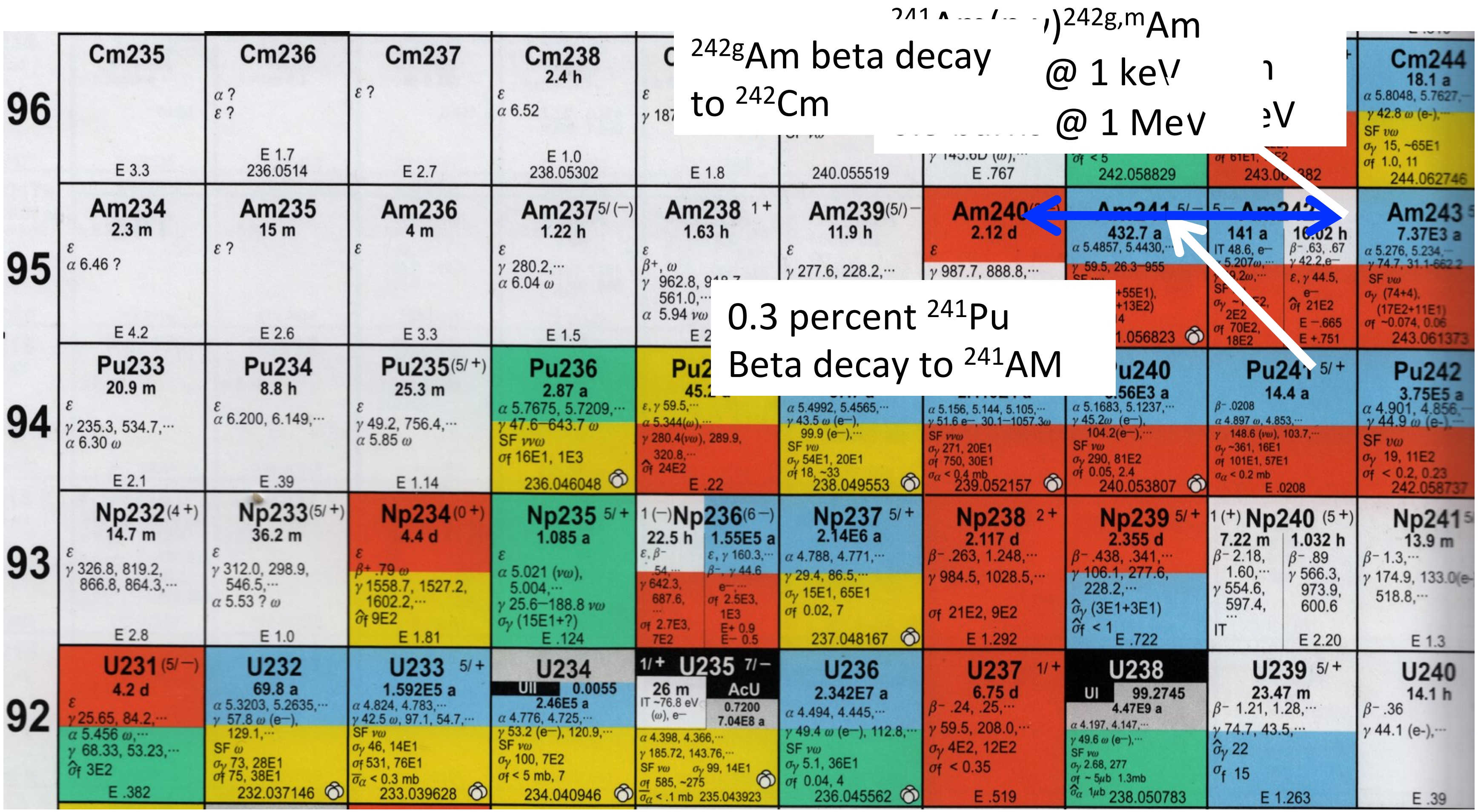
^{238}Pu(n,2n)^{237}Pu
Threshold 7.0 MeV
0.12 barn @ 14 MeV

8Pu
7 MeV
.4 MeV

^{240}Pu(n,γ)^{241}Pu
3 barns @ 1 keV

Depletion by fission,
Threshold fission for even masses.

Neutron Reactions with the Actinides
(Americium)



Actinide Isotopes in Nuclear Debris

- The equations for the successive reactions are solved numerically in a code using the Runga-Kutta method.
- The isotopes produced give the neutron fluence and spectrum that the original material was exposed to.

The Advantage of Isotope Ratios

If we assume no burnup the first order equation is

$$\frac{dN_{A-1}}{dt} = \sigma_1 N_A \phi. \tag{1}$$

Assume constant flux, the solution is

$$N_{A-1} = \sigma_1 N_A \phi t. \tag{2}$$

The second order reaction is then

$$\frac{dN_{A-2}}{dt} = \sigma_2 \left[\sigma_1 N_A \phi t\right] \phi. \tag{3}$$

The solution is

$$\begin{aligned} N_{A-2} &= \sigma_1 \sigma_2 N_A \int_0^t \phi^2 t dt \\ &= \frac{\sigma_1 \sigma_2 N_A (\phi t)^2}{2}. \end{aligned} \tag{4}$$

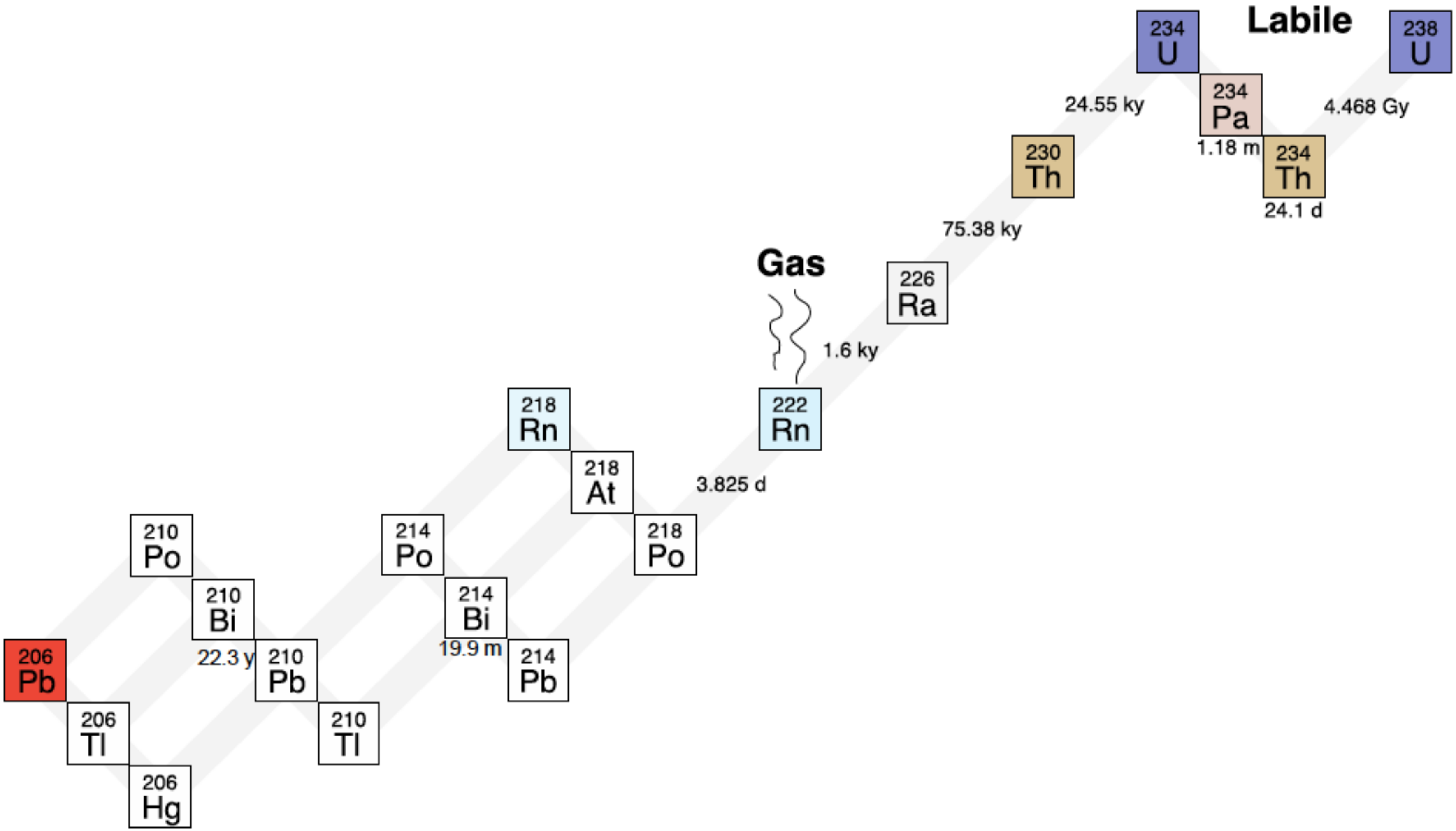
Therefore the atom ratio is

$$\frac{N_{A-2}}{N_{A-1}} = \frac{\sigma_2}{2} \phi t. \tag{5}$$

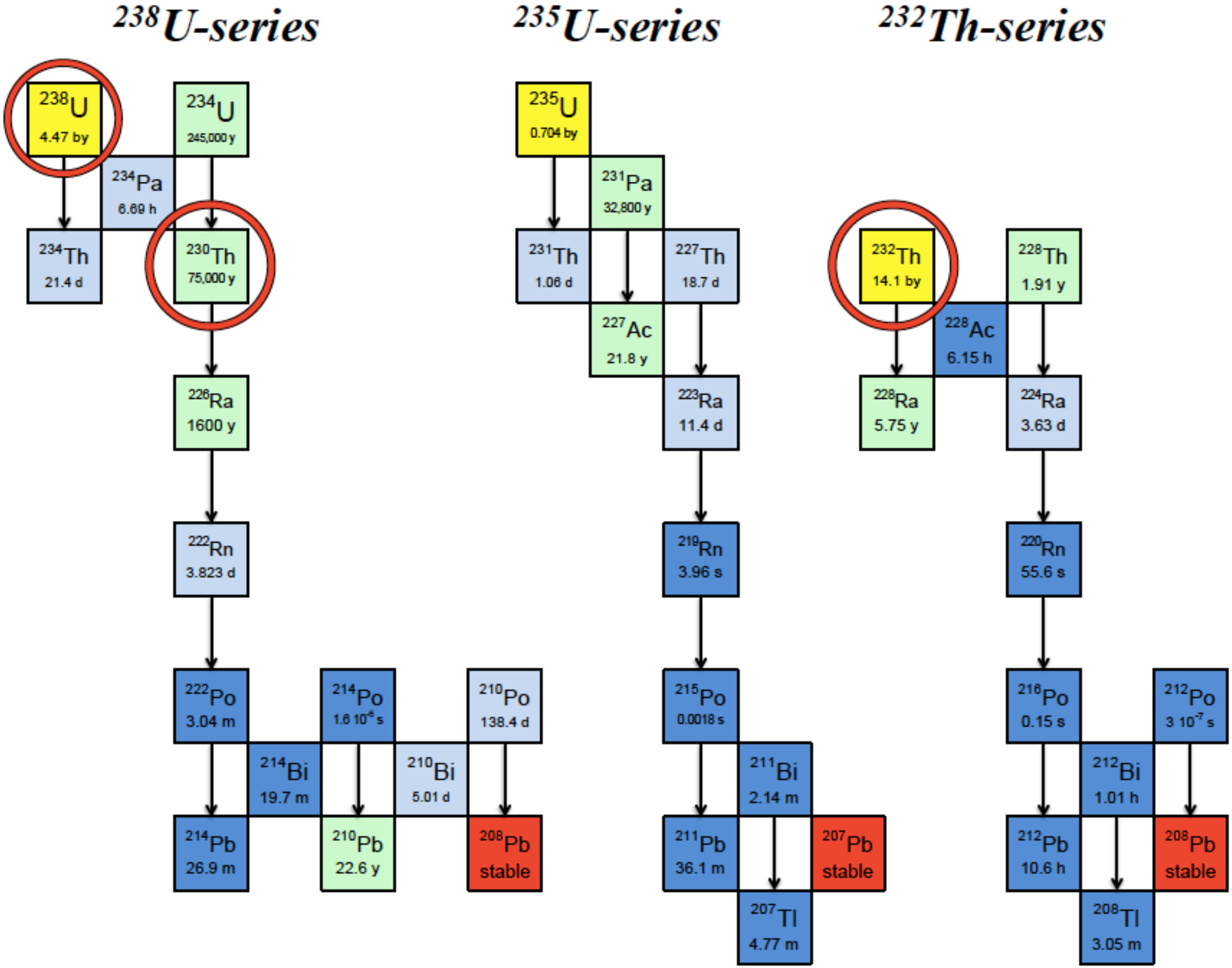
Growth and Decay

- The growth and decay of radioactive isotopes are similar to the transmutation of isotopes in high flux environment.
- The Bateman equations are useful. But restricted to specific initial conditions.
- The Bateman equations can be modified to include transmutation.

^{238}U Natural Decay Series



Peucker-Ehrenbrink, 2012



Peucker-Ehrenbrink, 2012

Bateman Equations

Start with a simple parent/daughter growth and decay. The equation for the daughter is,

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2$$

or

$$\frac{dN_2}{dt} + \lambda_2 N_2 - \lambda_1 N_1^0 e^{-\lambda_1} = 0. \qquad (1)$$

The solution of this linear differential equation of the first order may be obtained by standard methods and gives

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 \left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) + N_2^0 e^{-\lambda_2 t}. \qquad (2)$$

Bateman Equations

Consider the grandaughter. The equation for its growth and decay is,

$$\frac{dN_3}{dt} = \lambda_2 N_2 - \lambda_3 N_3. \tag{3}$$

Eq. 3 is analgous to Eq. 1, but the solution calls for more labor, because N_2 is a much more complicated function than N_1. The great grandaughter is even more complicated. Fortunately, H. Bateman has given the solution for a chain of n members with the special assumption that at $t = 0$ the parent substance alone is present, that is, $N_2^0 = N_3^0 = \cdots = N_n^0 = 0$. This solution is

$$\begin{aligned} N_n &= C_1 e^{-\lambda_1 t} + C_2 e^{-\lambda_2 t} + \cdots C_n e^{-\lambda_n t}, \\ C_1 &= \frac{\lambda_1 \lambda_2 \cdots \lambda_{n-1}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)\cdots(\lambda_n - \lambda_1)} N_1^0, \\ C_2 &= \frac{\lambda_1 \lambda_2 \cdots \lambda_{n-1}}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)\cdots(\lambda_n - \lambda_2)} N_1^0, \text{ and so on.} \end{aligned} \tag{4}$$

Bateman Applied to Transmutation

Successive neutron reactions in a high flux, such as, a high flux reactor can be solved using the Bateman equations, as well. The rate of disappearence of an isotope in a neutron flux is

$$-\frac{dN}{dt} = (\lambda + n\nu\sigma)N = \Lambda N. \tag{5}$$

Consider a parent daughter pair. The parent disappears by both transmutation and decay. But the daughter grows by decay of the parent only and disappears by both processes. In general notation,

$$\frac{dN_{i+1}}{dt} = \lambda_i N_i - \Lambda_{i+1} N_{i+1}.$$

We replace λ_i by a modified decay constant $\Lambda_i^* = \lambda_i^* + n\nu\sigma_i^*$, were only the decay constant of transmuation term that lead to next progeny in the chain is used.

Bateman Applied to Transmutation

With this nomenclature the Bateman equation becomes,

$$
\begin{aligned}
N_n &= C_1 e^{-\Lambda_1 t} + C_2 e^{-\Lambda_2 t} + \cdots C_n e^{-\Lambda_n t}, \\
C_1 &= \frac{\Lambda_1^* \Lambda_2^* \cdots \Lambda_{n-1}^*}{(\Lambda_2 - \Lambda_1)(\Lambda_3 - \Lambda_1)\cdots(\Lambda_n - \Lambda_1)} N_1^0, \\
C_2 &= \frac{\Lambda_1^* \Lambda_2^* \cdots \Lambda_{n-1}^*}{(\Lambda_1 - \Lambda_2)(\Lambda_3 - \Lambda_2)\cdots(\Lambda_n - \Lambda_2)} N_1^0, \text{and so on.}
\end{aligned}
\tag{6}
$$

An Example

As an illustration, we compute the amount of 3.15-d ^{199}Au formed by two successive (n, γ) reactions when 1 g ^{197}Au is exposed for 30 h in a neutron flux of 1×10^{14} cm^{-2} s^{-1}. The chain of reactions is

$$^{197}\mathrm{Au} \xrightarrow[n,\,\gamma]{\sigma=99\mathrm{b}} {}^{198}\mathrm{Au} \xrightarrow[n,\,\gamma]{\sigma=2.5\times 10^4\mathrm{b}} {}^{199}\mathrm{Au}$$

$$^{198}\mathrm{Au}: \ \beta^- \downarrow \ t_{1/2}=2.70\ \mathrm{d} \qquad {}^{199}\mathrm{Au}: \ \beta^- \downarrow \ t_{1/2}=3.14\ \mathrm{d}$$

We use (5-12) for this three-membered chain:

$$N_{199} = \Lambda^*_{197}\Lambda^*_{198}N^0_{197}\left[\frac{e^{-\Lambda_{197}t}}{(\Lambda_{198}-\Lambda_{197})(\Lambda_{199}-\Lambda_{197})} + \frac{e^{-\Lambda_{198}t}}{(\Lambda_{197}-\Lambda_{198})(\Lambda_{199}-\Lambda_{198})} + \frac{e^{-\Lambda_{199}t}}{(\Lambda_{197}-\Lambda_{199})(\Lambda_{198}-\Lambda_{199})}\right].$$

The numerical values to be substituted are

$$t = 1.08 \times 10^5 \text{ s},$$

$$nv = 10^{14} \text{ cm}^{-2} \text{ s}^{-1},$$

$$\sigma_{197} = 9.9 \times 10^{-23} \text{ cm}^2,$$

$$\sigma_{198} = 2.5 \times 10^{-20} \text{ cm}^2,$$

$$N_{197}^0 = \frac{6.02 \times 10^{23}}{197} = 3.05 \times 10^{21},$$

$$\Lambda_{197}^* = \Lambda_{197} = nv\sigma_{197} = 9.9 \times 10^{-9} \text{ s}^{-1},$$

$$\begin{aligned}\Lambda_{198} &= \lambda_{198} + nv\sigma_{198} = 3.0 \times 10^{-6} + 2.5 \times 10^{-6} \\ &= 5.5 \times 10^{-6} \text{ s}^{-1},\end{aligned}$$

$$\Lambda_{198}^* = nv\sigma_{198} = 2.5 \times 10^{-6} \text{ s}^{-1},$$

and

$$\Lambda_{199} = \lambda_{199} = 2.55 \times 10^{-6} \text{ s}^{-1}.$$

Using these values, we get

$$N_{199} = 7.85 \times 10^7 \left(\frac{e^{-0.00107}}{5.5 \times 10^{-6} \times 2.55 \times 10^{-6}} + \frac{e^{-0.594}}{5.5 \times 10^{-6} \times 2.95 \times 10^{-6}} - \frac{e^{-0.275}}{2.55 \times 10^{-6} \times 2.95 \times 10^{-6}} \right)$$

$$= 7.55 \times 10^7 (7.12 \times 10^{10} + 3.40 \times 10^{10} - 1.01 \times 10^{11}) = 3.2 \times 10^{17}.$$

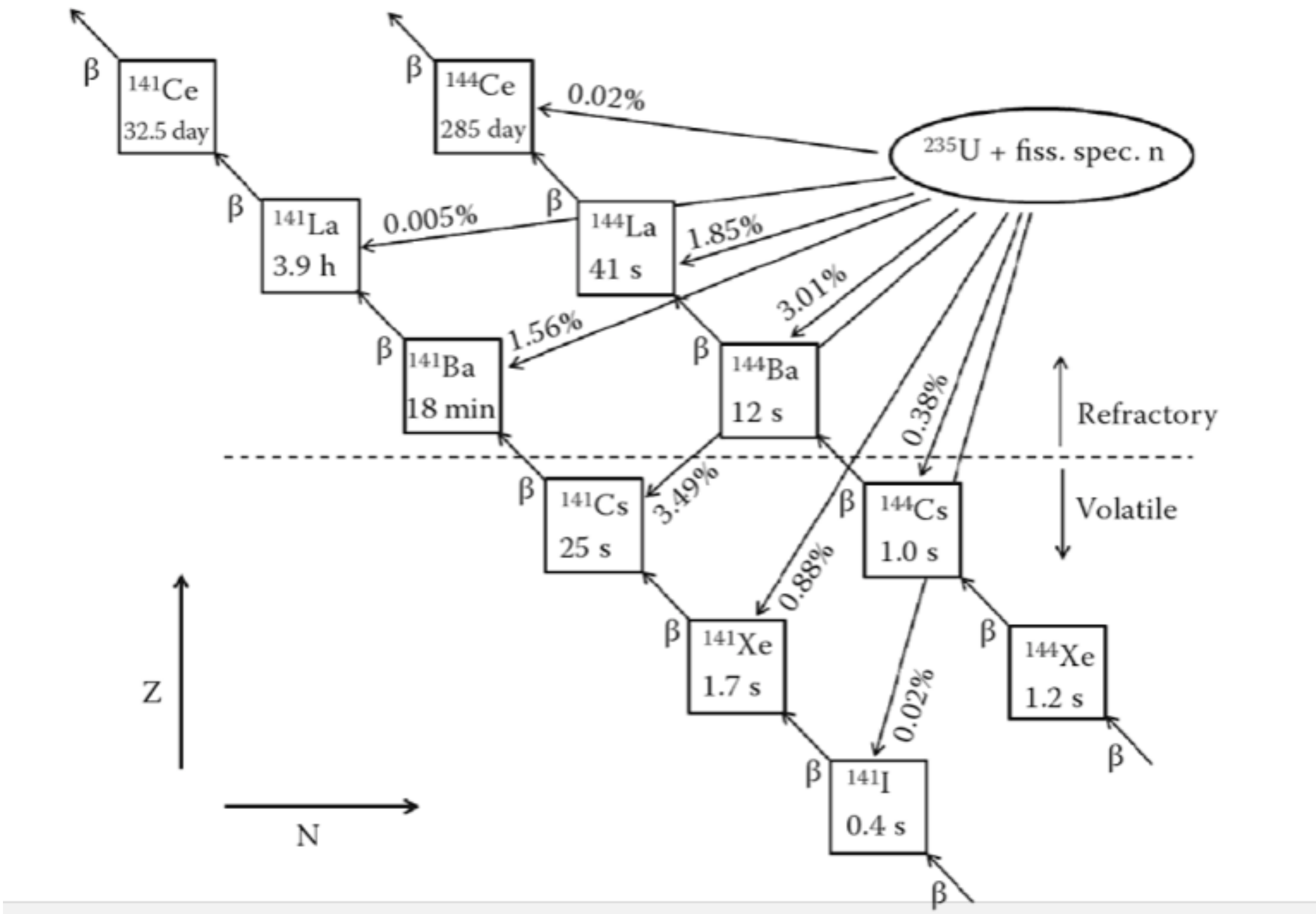
The disintegration rate of ^{199}Au at the end of the irradiation is $\lambda_{199} N_{199} = 0.82 \times 10^{12}\ \mathrm{s}^{-1}$. For comparison we compute the disintegration rate of ^{198}Au in the sample [again from (5-12) for a two-membered chain]:

$$\lambda_{198} N_{198} = \lambda_{198} n v \sigma_{197} N_{197}^0 \left(\frac{e^{-\Lambda_{197} t}}{\Lambda_{198} - \Lambda_{197}} + \frac{e^{-\Lambda_{198} t}}{\Lambda_{197} - \Lambda_{198}} \right)$$

$$= 9.06 \times 10^7 \frac{0.999 - 0.552}{5.5 \times 10^{-6}} = 7.36 \times 10^{12}\ \mathrm{s}^{-1}.$$

Thus about 10 percent of the radioactive disintegrations in the sample occur in ^{199}Au.

Fission Does Not Always Meet the Initial Conditions



The GD Code

The general differential equations for radioactive decay and growth

$$\begin{aligned}
\frac{dN_1}{dt} &= a_{11}N_1 && +r_1(t) \\
\frac{dN_2}{dt} &= a_{21}N_1 + a_{22}N_2 && +r_2(t) \\
&\vdots \\
\frac{dN_n}{dt} &= a_{n1}N_1 + \cdots + a_{nn}N_n && +r_n(t)
\end{aligned} \tag{1}$$

where $N_i(t)$ is the number of atoms of nuclide i existing at time t, a_{ij}, $i \geq j$ are constants, and r_i's are the rates of formation from sources other than by decay of isotopes i. A specialized form $r_i(t) = y_i f(t)$ is used in the Los Alamos code GD, so that the rates have time independent ratios to each other. The y_i's are called fractional independent yields.

GD Code Continued

where $\mathbf{A}$ is the matrix of a_{ij}'s, $\mathbf{N}$ and $\mathbf{Y}$ are column vectors and $f(t)$ is a scalar. Equation (2) has the solution

$$\mathbf{N}(t) = e^{(t-\tau)\mathbf{A}}\mathbf{N}(\tau) + \int_{\tau}^{t} e^{(t-s)\mathbf{A}} f(t)\mathbf{Y} ds \tag{3}$$

There is a non singular matrix $\mathbf{P}$ such that

$$\mathbf{P}^{-1}\mathbf{A}\mathbf{P} = \mathbf{J} \tag{4}$$

is diagonal. Th GD code finds $\mathbf{P}$. The solution is then

$$\begin{aligned} \mathbf{N}(t) &= \mathbf{P}\, diag\left[e^{a_{11}(t-\tau)}, \ldots, e^{a_{nn}(t-\tau)}\right] \mathbf{P}^{-1}\mathbf{N}(\tau) \\ &+ \mathbf{P}\left(\int_{\tau}^{t} diag\left[e^{a_{11}(t-s)}, \ldots, e^{a_{nn}(t-s)}\right] f(s) ds\right) \mathbf{P}^{-1}\mathbf{Y} \end{aligned} \tag{5}$$

The GD Code Solution

Define $F_i(t)$ by

$$F_i(t-\tau) = \int_\tau^t e^{a_{ii}(t-s)} f(s) ds \tag{6}$$

then the solution is

$$\begin{aligned} \mathbf{N}(t) &= \mathbf{P}\, diag\left[e^{a_{11}(t-\tau)}, \ldots, e^{a_{nn}(t-\tau)}\right] \mathbf{P}^{-1}\mathbf{N}(\tau) \\ &+ \mathbf{P}\, diag\left[F_1(t-\tau), \ldots, F_n(t-\tau)\right] \mathbf{P}^{-1}\mathbf{Y} \end{aligned} \tag{7}$$

The function $f(s)$ is taken to be piecewise constant with f_j on time intervals U_j to W_j. With this specialization it follows that

$$F_i(t-\tau) = \frac{e^{a_{ii}t}}{-a_{ii}} \sum_j f_j \left(e^{-a_{ii}W_j} - e^{-a_{ii}U_j}\right). \tag{8}$$